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(71)Applicant : TOYOTA CENTRAL RES & DEV LAB  
INC

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(72)Inventor : NONOYAMA FUMIO  
SUZUKI KENICHI  
ABE KATSUJI  
ASAOKA MASAHIKO

## (54) SEPARATOR FOR FUEL CELL

## (57)Abstract:

PROBLEM TO BE SOLVED: To provide a separator for a fuel cell excellent in current collecting performance in a use environment and offering no cost problem, by using a metallic material excellent in workability as a base material.

SOLUTION: A separator for a fuel cell constituting a solid high-polymer type fuel cell comprises a material made by applying a coating layer composed of Sn, an Sn-containing alloy, an Sn-containing compound, RuO<sub>2</sub>, IrO<sub>2</sub>, MnO<sub>2</sub>, or WC on a surface of a metallic material excellent in corrosion resistance. Because the metallic material is used as a base material, separator press forming workability is excellent and an oxide film formed on the Sn-containing coating layer during use is high in electroconductivity, and current collecting performance is prevented from lowering during use.

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**CLAIMS**

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**[Claim(s)]**

[Claim 1] Separator for fuel cells characterized by consisting of material which has the alloy which is the separator for fuel cells equipped with the current collection performance which contacts each electrode which pinches a solid electrolyte from both sides, is arranged, and forms the gas passageway of distributed gas between these electrodes, and contains Sn and Sn on the front face of the metallic material excellent in corrosion resistance, a compound containing Sn, and the enveloping layer which consists of RuO<sub>2</sub>, IrO<sub>2</sub>, MnO<sub>2</sub>, or WC.

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**DETAILED DESCRIPTION**

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**[Detailed Description of the Invention]**

**[0001]**

[The technical field to which invention belongs] this invention relates to the separator for fuel cells.

**[0002]**

[Description of the Prior Art] the composition whose separator for fuel cells constitutes a solid-state macromolecule type fuel cell -- it is a member, and each electrode which pinches a solid electrolyte from both sides is contacted, it is arranged, distributed gas paths, such as fuel gas and oxidizer gas, are formed between these electrodes, and it has the current collection performance which contacts an electrode and derives current

[0003] Generally this separator consists of metallic materials, such as precise carbon graphite, or SUS, Ti, Cu, aluminum, as shown in JP,4-95354,A, JP,8-222237,A, etc.

[0004]

[Problem(s) to be Solved by the Invention] By the way, since a high current collection performance is maintained also by prolonged use highly [ a current collection performance ], the separator which consists of precise carbon graphite can be called outstanding separator from a viewpoint of a current collection performance. However, many heights, a slot, etc. are formed in the field which counters the electrode of separator that the path plasticity ability for forming a gas passageway should be given. However, since precise carbon graphite is a very weak material, it has the problem that mass production is difficult while it is not easy to machine cutting etc. that many heights and slots should be formed in the front face of separator and processing cost becomes high.

[0005] On the other hand, since the metallic material is excellent in intensity and ductility in the above-mentioned separator which solves and consists of metallic materials as compared with precise carbon graphite, press working of sheet metal which forms many heights and slots that the path plasticity ability of a gas passageway should be given is possible, and there is an advantage that processing cost is low and mass production is also easy. However, under the operating environment of separator, an oxide film is generated by the front face, and it is [ a metallic material ] easy, and it has the problem that contact resistance with the electrode of the generated oxide film is large, and reduces the current collection performance of separator.

[0006] Adopting as the front face of the metallic material excellent in processability the material which coated carbon graphite excellent in electrical conductivity as a component of separator that these problems should be coped with is proposed. Moreover, it is possible to coat metallic materials, such as Au excellent in electrical conductivity. However, the material of the former which makes a metallic material a base material has a problem also in processing cost while reliance lacks in the airtightness of the coating material to a base material, and adhesion. Moreover, the latter material has a problem in the point that coating material is expensive.

[0007] Therefore, the purpose of this invention makes the metallic material excellent in processability a base material, and it excels in the electrical conductivity under an operating environment, and it is to offer the separator for fuel cells which is satisfactory in cost.

[0008]

[Means for Solving the Problem] this invention contacts the separator for fuel cells, and each electrode which pinches a solid electrolyte from both sides especially, is arranged, and form the gas passageway of distributed gas between these electrodes. It is the separator for fuel cells equipped with the current collection performance, and is characterized by consisting of material which has the alloy which contains Sn and Sn on the front face of the metallic material excellent in corrosion resistance, a compound containing Sn, and the enveloping layer which consists of RuO<sub>2</sub>, IrO<sub>2</sub>, MnO<sub>2</sub>, or WC.

[0009]

[Embodiments of the Invention] The separator concerning this invention can make a base material the metallic material excellent in corrosion resistance, and can mention SUS, Ti, Cu, aluminum, these alloys, etc. as this metallic material. Moreover, the enveloping layers which constitute the separator concerning this invention are the alloy containing Sn and Sn, a compound containing Sn, and RuO<sub>2</sub>, IrO<sub>2</sub>, MnO<sub>2</sub> or WC.

[0010] As an alloy containing Sn, Sn-nickel, Sn-Fe, Sn-Ti, etc. can be mentioned. Moreover, SnO<sub>2</sub> can be mentioned as a compound containing Sn. As an oxide, RuO<sub>2</sub>, IrO<sub>2</sub>, MnO<sub>2</sub>, WC, etc. can be mentioned to others. These may be used together even if independent. Generally, although the enveloping layer which constitutes the separator concerning this invention coats each above-mentioned material on the surface of a base material and is formed, it precedes coating such material and can coat other materials on the surface of a base material. For example, it precedes coating Sn on the surface of a base material, and nickel can be beforehand coated on the surface of a base material. As a coating means, meanses, such as plating, \*\*\*\*\*, ion plating, and an application, are employable.

[0011] Moreover, in the separator concerning this invention, after carrying out press forming of heating at the temperature beyond the melting point of Sn after forming an enveloping layer on the surface of a base material, and the material in which the enveloping layer was formed on the front face of a base material to the configuration of separator, it can heat at the temperature beyond the melting point of Sn. After carrying out press forming of the material which formed the enveloping layer in both sides of a base material in the separator concerning this invention to the configuration of separator, it can consider as the structure of joining both separator mutually through Sn and having a channel for cooling among these both, a fuel gas side further again by carrying out the polymerization of the oxidizer gas side separator, and heating at the temperature beyond the melting point of Sn.

[0012]

[Function and Effect of the Invention] In the separator for fuel cells which this invention makes applicable to application, since the contact section with an electrode is oxidized by the oxidizing atmosphere which a hot steam forms at the time of operation of a fuel cell when this separator consists only of metallic materials, such as SUS, Ti, Cu, and aluminum, an oxide film is formed in the contact part concerned. For this reason, separator makes contact resistance increase and makes current collection efficiency fall.

[0013] On the other hand, the separator concerning this invention makes a corrosion resistance metallic material a base material, and is equipped with the alloy which contains Sn and Sn on the front face, the compound containing Sn, and the enveloping layer which consists of RuO<sub>2</sub>, IrO<sub>2</sub>, MnO<sub>2</sub>, or WC. Even if the alloy containing Sn and Sn has high electrical conductivity in itself, and oxidizes even if and it becomes an oxide, since the fall of electrical conductivity is very small, making contact resistance increase by a fuel cell being on stream does not almost have it, and it holds high current collection efficiency. Moreover, the compound containing Sn, and RuO<sub>2</sub>, IrO<sub>2</sub>, MnO<sub>2</sub> and WC have high electrical conductivity in the state where it already oxidized. For this reason, there is almost no making contact resistance increase by a fuel cell being on stream, and it holds high current collection efficiency.

[0014] The alloy containing Sn and Sn or the compound containing Sn, and RuO<sub>2</sub>, IrO<sub>2</sub>, MnO<sub>2</sub> and WC can be coated with meanses, such as plating, electrolytic plating, ion plating, and an application, easily and cheaply to the front face of a base material. Moreover, become empty and there is no possibility that the enveloping layer of airtightness and adhesion formed of coating may be good, and may exfoliate from a base-material front face.

[0015]

[Test Example(s)] the separator which forms the component of the separator which starts this invention to know the existence of the influence to the contact resistance at the time of being exposed to steam atmosphere, and starts this invention about the sheet metal with which this invention consists of various kinds of metallic materials which constitute the separator for fuel cells made applicable to application — composition — the solid-state macromolecule type fuel cell used as a member was constituted, and examination about the cell property was performed

Ti which is the metallic material currently generally used as separator in the example of an exam, (Example 1 of an examination) The sheet metal which consists of aluminum, Cu, SUS316, and SUS304, the sheet metal which comes to coat the front face of the sheet metal made from SUS304 carbon graphite, and the board which

consists of precise carbon graphite are produced, respectively. These sheet metal and boards were exposed to the steam atmosphere of 100% of humidity at 80 degrees C for 96 hours, and the contact resistance before and after exposure in the steam atmosphere in these sheet metal and boards was measured. The obtained result is shown in the graph of drawing 1 .

[0016] Although it is comparatively small before exposing contact resistance to a steam in the sheet metal which consists of Ti, aluminum, Cu, SUS316, and SUS304 when the result of drawing 1 is referred to, increasing remarkably will be admitted if exposed to steam atmosphere. Since a steam is an oxidizing atmosphere, an oxide film is generated by the sheet metal front face, and this is understood to be what originates in generation of an oxide film and contact resistance is increasing sharply. Therefore, in the separator which makes these sheet metal a component, contact resistance increases on stream [ a fuel cell ], and current collection efficiency falls remarkably.

[0017] On the other hand, with the sheet metal which comes to coat the front face of the sheet metal made from SUS304 carbon graphite, and the board which consists of precise carbon graphite, hardly changing, even if it is small before exposing contact resistance to steam atmosphere, and exposed to steam atmosphere is admitted. Therefore, in the separator which makes these sheet metal a component, current collection efficiency there are few increases in contact resistance also during operation of a fuel cell, and high is held.

(Example 2 of an examination) In the example of an exam, the sheet metal of SUS304 which is the metallic material currently generally used as separator is made into the base material. And the front face of this base material is coated with Sn by the cladding, and it is formed in it. The thickness of the enveloping layer of Sn component produced sheet metal as 30 micrometers, 20 micrometers, and 10 micrometers, respectively. These sheet metal was exposed to the steam atmosphere of 100% of humidity at 80 degrees C for 96 hours, and the contact resistance before and after exposure in the steam atmosphere in these sheet metal was measured. The obtained result is shown in the graph of drawing 2 .

[0018] Reference of the result of drawing 2 admits that the contact resistance before exposing to steam atmosphere is remarkably small as compared with the sheet metal which consists of Ti, aluminum, Cu, SUS316, and SUS304, and the increase in the contact resistance after being exposed to steam atmosphere is very small in the sheet metal which has the enveloping layer of Sn component. Therefore, in the separator which makes these sheet metal a component, current collection efficiency

there are few increases in contact resistance also during operation of a fuel cell, and high is held.

[0019] In addition, in the example of an exam, the sheet metal which has an enveloping layer with a thickness of 2 micrometers – 10 micrometers in which it excels in corrosion resistance on the front face of the base material which is sheet metal of SUS304, and electrical conductivity comes to coat SnO<sub>2</sub>, RuO<sub>2</sub>, IrO<sub>2</sub>, MnO<sub>2</sub> [ high ], and high WC by ion plating, respectively was produced, respectively. And when the examination exposed to the same steam atmosphere as the above was tried, it was admitted not to mention the contact resistance before exposing any sheet metal to steam atmosphere being remarkably small that the increase in the contact resistance after exposure in steam atmosphere was very small.

In the example of an exam, the sheet metal of SUS304 which is the metallic material currently generally used as separator is made into a base material. (Example 3 of an examination) Come to coat the front face of this base material Sn by the cladding. The sheet metal which has the enveloping layer of Sn component (30 micrometers in thickness, 20 micrometers, and 10 micrometers), And after giving nickel plating with a thickness of 2 micrometers for the sheet metal of SUS304 as a base material on the surface of *Perilla frutescens* (L.) Britton var. *crispa* (Thunb.) Decne., the sheet metal which has the enveloping layer of Sn component (30 micrometers in thickness which comes to coat the front face of nickel plating layer Sn by the cladding, 20 micrometers, and 10 micrometers) was produced, respectively. And these sheet metal was heat-treated at 250 degrees C by the non-oxidizing atmosphere for 1 hour.

[0020] Each heat-treated sheet metal was exposed to the steam atmosphere of 100% of humidity at 80 degrees C for 96 hours, and the contact resistance before and after exposure in the steam atmosphere in these sheet metal was measured. The obtained result is shown in the graph of drawing 3 . If these results are referred to, as compared with the sheet metal which shows the contact resistance in these sheet metal to the example 2 which has the enveloping layer of Sn component, it will be lower than it and it will be admitted that the increase in the contact resistance after being exposed to steam atmosphere is equivalent or very small. Therefore, in the separator which makes these sheet metal a component, current collection efficiency there are few increases in contact resistance also during operation of a fuel cell, and high is held.

[0021] It is admitted [ in / the sheet metal which comes to form the enveloping layer of Sn component with a thickness of 10 micrometers in the front face of nickel plating layer after giving nickel plating with a thickness of 2 micrometers especially to the front face of the sheet metal-like base material of SUS304 / both ] that contact

resistance equivalent to the precise carbon graphite shown in the example 1 of an examination before and after exposure in steam atmosphere is shown. In addition, although the above-mentioned heat-treatment is performed in the non-oxidizing atmosphere, you may heat-treat by the usual oxidizing atmosphere. In this case, since an oxide film is generated by the front face of the enveloping layer which covers a base material, it is desirable to remove the generated oxide film.

The sheet metal which comes to form the enveloping layer of Sn component with a thickness of 10 micrometers in the front face of the sheet metal-like base material of SUS304 in the example of an exam, (Example 4 of an examination) After giving nickel plating with a thickness of 2 micrometers to the front face of the sheet metal-like base material of SUS304, on the front face of nickel plating layer 10 micrometers in and thickness About what heat-treated the sheet metal which comes to form the enveloping layer of 30-micrometer Sn component at 250 degrees C by the non-oxidizing atmosphere for 1 hour, change of the cross-section organization of an enveloping layer and the X-ray diffraction of an enveloping layer were performed. The result is shown in drawing 4 - drawing 8 .

[0022] Drawing 4 shows the optical microscope photograph of the cross-section organization before and behind heat-treatment of the sheet metal which comes to form the enveloping layer of Sn component with a thickness of 10 micrometers in the front face of the sheet metal-like base material of SUS304. Drawing 5 shows the chart of the X-ray diffraction of the enveloping layer after heat-treatment of this sheet metal. It was checked that it is a Sn-Fe compound as a result of a clear organization change being accepted by heat-treatment at 250 degrees C and analyzing this organization, when this cross-section organization was referred to.

[0023] Drawing 6 shows the optical microscope photograph of the cross-section organization before and behind heat-treatment of the sheet metal which comes to form the enveloping layer of Sn component with a thickness of 10 micrometers in the front face of nickel plating layer, after giving nickel plating with a thickness of 2 micrometers to the front face of the sheet metal-like base material of SUS304. Drawing 7 shows the chart of the X-ray diffraction of the enveloping layer after heat-treatment of this sheet metal. It was checked that it is a Sn-nickel compound as a result of a clear organization change being accepted by heat-treatment at 250 degrees C and analyzing this organization, when this cross-section organization was referred to.

[0024] Drawing 8 shows the optical microscope photograph of the cross-section organization after heat-treatment of the sheet metal which comes to form the



enveloping layer of Sn component with a thickness of 30 micrometers in the front face of nickel plating layer, after giving nickel plating with a thickness of 2 micrometers to the front face of the sheet metal-like base material of SUS304. As a result of analyzing this enveloping layer, it was checked that the Sn-nickel compound is generating to inside at the enveloping layer. However, it is checked that Sn remains on the surface of an organization, and the contact resistance of the sheet metal after this heat-treatment originates in the surface remains Sn, and is accepted to be a thing of the same grade as the sheet metal before heat-treatment. Therefore, if there are too many amounts of coating of Sn, even if it heat-treats, Sn remains on a front face and the effect of heat-treatment cannot be expected.

[0025] It heats, when forming the enveloping layer of Sn component in the front face of nickel layer from the above result after coating the front face of the sheet metal-like base material of SUS304 with nickel. In this case, as for nickel layer, it is desirable that it is the range whose thickness is 2 micrometers – 10 micrometers. When nickel layer thickness is less than 2 micrometers, the rate of Sn which remains on a front face becomes high after heat-treatment, and the effect of heat-treatment cannot be expected. Moreover, in case press forming is carried out to the configuration of separator while it is disadvantageous on cost when nickel layer thickness exceeds 10 micrometers, there is a possibility of producing trouble.

[0026] In addition, when changing to the sheet metal-like base material of SUS and adopting metallic materials, such as Ti, it is the same, and when forming the enveloping layer of Sn component in the front face of the sheet metal-like base material of SUS304, it heats. Because, in 5 micrometers or more, since the rate of Sn which remains after heat-treatment increases, the property of lowering the contact resistance of the generated Sn-Ti compound is not fully harnessed.

(Example 5 of an examination) In the example of an exam, press forming of the sheet metal which formed the enveloping layer of Sn component in the front face of nickel layer after coating the front face or this front face of a sheet metal-like base material (0.1mm – 0.3mm in thickness) of SUS304 and Ti with nickel was carried out to the configuration of the separator for fuel cells so that it might become the electrode contact side whose enveloping layer side has much salients. Subsequently, the sheet metal which carried out press forming was heat-treated at the temperature beyond the melting point of Sn under the non-oxidizing atmosphere, and the separator for fuel cells was formed.

[0027] It was checked by the obtained separator that the Sn-Fe compound, the Sn-Ti compound, the Sn-nickel compound, etc. exist in the front face by the side of

electrode contact. in addition, the usual fabrication whose above-mentioned press forming has vertical both molds -- the forming means which comes out enough, and exists with a forming means to use metal mold, and carries out hydraulic forming using a punch or female mold is sufficient In press forming, since the enveloping layer of Sn component is rich in \*\*\*\*\*, this enveloping layer is wearing the front face of a base material also [ near the severe height of deformation ], and the ablation from a base-material front face is not accepted.

(Example 6 of an examination) In the example of an exam, after coating both sides or these both sides of a sheet metal-like base material (0.1mm – 0.3mm in thickness) of SUS304 and Ti with nickel, press forming of both sides of the sheet metal in which the enveloping layer of Sn component was formed on the front face of nickel layer was carried out to the configuration of the separator for fuel cells so that it might become the electrode contact side which has much salients, and the sheet metal for the object for fuel gas side separator and oxidizer gas side separator Subsequently, the polymerization of both [ these ] the sheet metal was carried out, it heat-treated at the temperature beyond the melting point of Sn under the non-oxidizing atmosphere, and the fuel gas side separator to which both sheet metal of each other was joined through Sn, and oxidizer gas side separator formed the separator of one.

[0028] In the obtained separator, it can constitute in the structure of having a channel for cooling between the joints of both sheet metal, and reduction of the cost of the separator for fuel cells and conductive improvement between a fuel gas side and oxidizer gas side separator can be aimed at by considering as this composition. In addition, in this separator, it was checked that it is checked that the Sn-Fe compound, the Sn-Ti compound, the Sn-nickel compound, etc. exist in the front face by the side of electrode contact, and the enveloping layer has not exfoliated from a base-material front face.

(Example 7 of an examination) In the example of an exam, after giving nickel plating with a thickness of 2 micrometers to the front face of the sheet metal-like base material (0.15mm in thickness) of SUS304, press forming of the sheet metal in which the enveloping layer of Sn component with a thickness of 10 micrometers was formed on the front face of nickel layer was carried out, and it heat-treated at the temperature beyond the melting point of Sn under the non-oxidizing atmosphere after that, and fuel gas side separator and oxidizer gas side separator were formed. Both [ these ] separator is adopted, a solid-state macromolecule type fuel cell is constituted, the cell property is measured, and the result is shown in the graph of drawing 9 .

[0029] in addition, various kinds of separator which comes to carry out press forming of the sheet metal in which the enveloping layer of a precise carbon graphite component with a thickness of 10 micrometers was formed on the front face of the sheet metal-like base material (0.15mm in thickness) of SUS304, and the sheet metal of only the sheet metal-like base material (0.15mm in thickness) of SUS316 and Ti as an example of comparison -- composition -- the property of the solid-state macromolecule type fuel cell used as a member is collectively shown in the graph of drawing 9

[0030] When this drawing was referred to, in the separator for fuel cells concerning the example of an exam, it was admitted that a property almost equivalent to the separator made from precise carbon graphite was acquired, and, moreover, it was checked that this property does not almost have aging.

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[Translation done.]